Reinforcement vs strain-amplification in filled elastomers

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Abstract

Several mechanisms have been proposed to explain the remarkable mechanical properties of elastomers filled with sub-micrometric particles. The complexity of their structure and dynamics has been evidenced through many experimental techniques. Understanding and discriminating the various reinforcement mechanisms is a key issue in the field. We shall present an new approach combining different, complementary techniques, namely measurements of the mechanical response in conjunction with measurements of chain segment orientation by X-ray scattering, in which the response of the elastomer matrix in a composite material is selectively obtained [1, 2, 3, 4]. Crosslink densities are measured independently by multiple-quantum proton NMR. In unfilled materials, all measurements are nicely correlated, in agreement with rubber elasticity theory [1]. In filled materials, strong deviations with respect to the behavior of the pure unfilled elastomer are observed. The selective response of the elastomer matrix (as measured by X-ray scattering) gives access to chain over-stretching, that is, local strain amplification in the elastomer matrix, which may be qualified as the entropic contribution to reinforcement. This effect only contributes to a fraction of the overall reinforcement. This fraction is small in many cases of interest, namely in strongly reinforced materials (see figure 1). In that case, rigid network effects, which may be qualified as non-entropic contribution to reinforcement, are largely predominant. The relative contributions of each type of mechanism are quantified as a function of temperature, filler fraction and strain amplitude, in materials filled with precipitated silica. Associated physical phenomena will be discussed.
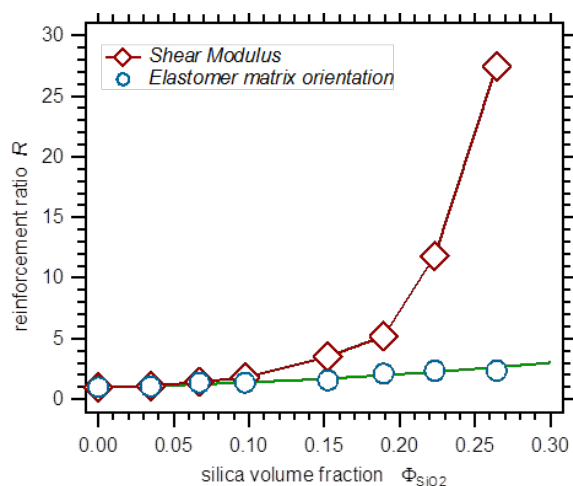
FIGURE 1 – Variation of the shear modulus and of the elastomer matrix average orientation induced upon stretching, vs the filler volume fraction in a series of silica-filled SBR elastomers. The elastomer matrix orientation gives the magnitude of the strain amplification effect, which is small comared to overall reinforcement in strongly reinforced samples.

Références

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